

Double quantum dot as a minimal thermoelectric generator

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Based on numerical renormalization group calculations, we demonstrate that experimentally realized double quantum dots constitute a minimal thermoelectric generator. In the Kondo regime, one quantum dot acts as an *n*-type and the other one as a *p*-type thermoelectric device. Properly connected, a capacitively coupled double quantum dot provides a miniature power supply utilizing the thermal energy of the environment.

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I. INTRODUCTION

The theoretical prediction [1] and subsequent experimental confirmation [2] of the Kondo effect in a quantum dot represents, without doubt, a prominent research highlight in the field of nanoscopic physics. While the Kondo effect of impurities in bulk metals induces a reduction of the conductance [3], its nanoscopic realization leads to an enhancement due to quantum many-body effects. More recently, the Kondo effect was also observed in double quantum dots [4–7]. Here, the two quantum dots may act as a pseudospin in analogy to spin-up and -down for the usual Kondo effect in a single quantum dot. Combined, spin and pseudospin can give rise to an enhanced SU(4) Kondo effect. Such double quantum dots are particularly exciting from the fundamental research point of view since (pseudo-)spin-up and -down can be controlled separately and their conductance can be measured independently [8].

Besides, for their electrical conductance, quantum dots are also considered as potential solid-state energy converters [9–12]. The high degree of tunability of nanoscale devices allows them to be operated at optimal thermoelectric efficiency. Promising in this respect are ultrasharp resonances, which can be achieved through the Kondo effect [13–17]. However, for the single-dot Kondo effect, the resonance is centered around the Fermi level so that electron and hole contributions cancel. The thermopower is vanishingly small [14]. A possibility to move the resonance away from the Fermi level is applying an external magnetic field. This splits the Kondo resonance with one spin-species above and the other below the Fermi energy, but the total (spin-averaged) thermopower stays small [15,16]. Another idea has been to employ the charge Kondo effect, realized in an Anderson impurity model with attractive interaction [15]. This is, however, difficult to realize experimentally. It requires, e.g., a strong electron-phonon coupling to realize an effective interaction that is attractive.

In this paper, we show that a capacitively coupled double quantum dot in the Kondo regime ultimately overcomes these difficulties. Such double dots, which are already realized experimentally, represent a stand-alone thermoelectric generator; see Fig. 1. Given an external heat gradient, (quasi-)electrons and holes dominate the linear transport in the two respective quantum dots such that a total current is generated if the quantum dots are suitably connected. We stress that the same theoretical concept can be realized in experimentally very distinct systems, such as molecular transport [18] and cold atoms [19].

II. MODEL AND METHOD

Basis of our calculations is the experimentally realized double quantum dot of Ref. [8]. The experimental signatures of the Kondo physics in the conductance are well described by an Anderson impurity model [6] with a single spin-degenerate level in each dot:

$$\begin{aligned} \hat{H} = & \sum_{i \in \{u,d\}} (\varepsilon_i \hat{n}_i + U_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}) + U' \hat{n}_u \hat{n}_d \\ & + \sum_{Ri,k,\sigma} \varepsilon_k \hat{c}_{Ri,k,\sigma}^\dagger \hat{c}_{Ri,k,\sigma} + \sum_{Ri,k,\sigma} (V_{Ri} \hat{a}_{i,\sigma}^\dagger \hat{c}_{Ri,k,\sigma} + \text{H.c.}). \end{aligned} \quad (1)$$

Here, $\hat{n}_{i,\sigma} = \hat{a}_{i,\sigma}^\dagger \hat{a}_{i,\sigma}$, where $\hat{a}_{i,\sigma}^\dagger$ ($\hat{a}_{i,\sigma}$) creates (annihilates) an electron with spin $\sigma \in \{\downarrow, \uparrow\}$ in dot $i \in \{u,d\}$ with energy level ε_i . U_i is the Coulomb interaction on quantum dot i (for simplicity we use $U_i = U$ in the following), and U' is the capacitive coupling between the two dots. Note that there is no direct tunneling between the two dots. $\hat{c}_{Ri,k,\sigma}^\dagger$ is the creation operator for an electron in the source or drain lead $R \in \{S,D\}$ with wavenumber k and energy ε_k . The leads are coupled to quantum dot i by the hybridization V_{Ri} , which corresponds to a tunneling rate $\Gamma_{Ri} = 2\pi\rho|V_{Ri}|^2$ for a constant lead density of states ρ (which can be taken as lead independent since only the combination $\rho|V_{Ri}|^2$ is of relevance).

As in Ref. [6], the numerical renormalization group (NRG) [20] approach is used to calculate the spectral function $A_i(\omega)$ of the two quantum dots from which the conductance G_i , the thermopower S_i , and the electronic contribution to the thermal conductance K_i^e can be determined in linear response via the Meir-Wingreen formula [21]; see Ref. [14]:

$$G_i(T) = e^2 I_i^0(T) \quad (2)$$

$$S_i(T) = -\frac{1}{|e|T} \frac{I_i^1(T)}{I_i^0(T)} \quad (3)$$

$$K_i^e(T) = \frac{1}{T} \left[I_i^2(T) - \frac{I_i^1(T)^2}{I_i^0(T)} \right]. \quad (4)$$

Here, e is the elementary charge, T the temperature, and I_i^n denote the transport integrals

$$I_i^n(T) = \frac{2}{h} \int d\omega \omega^n T_i(\omega) \left(-\frac{\partial f(\omega)}{\partial \omega} \right), \quad (5)$$

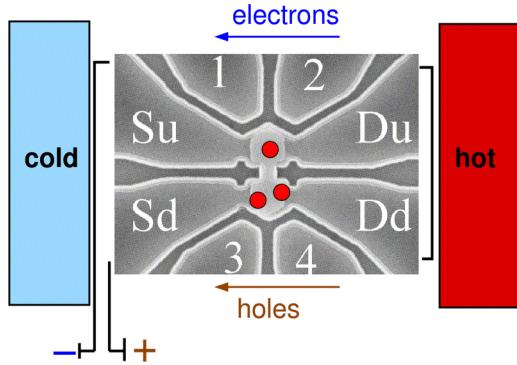


FIG. 1. (Color online) Schematic figure of the considered thermoelectric double quantum dot device. For suitable gate voltages of the electrodes 1,2 and 3,4, a negative current of (quasi-)electrons flows in the up quantum dot and a positive current (quasi-)holes in the down quantum dot. Hence, if the two dots are connected on the right-hand side, the up and down quantum dots provide on the left-hand side the negative and positive pole of a power supply energized by excess heat from the environment (the micrograph of the quantum dot at the center has been reproduced from Ref. [8]).

where the transmission is given by

$$\mathcal{T}_i(\omega) = 2\pi \frac{\Gamma_{Li}\Gamma_{Ri}}{\Gamma_{Li} + \Gamma_{Ri}} A_i(\omega). \quad (6)$$

The I_i^n are thus the moments of the spectral function weighted by the derivative of the Fermi function $f(\omega)$ around the Fermi energy $\omega = 0$.

III. GENERAL DISCUSSION

For a better understanding, we consider the Sommerfeld expansion of the thermopower at low T :

$$S_i(T) = -\frac{\pi^2 k_B}{3|e|} k_B T \frac{dA_i(\omega)/d\omega|_{\omega=0}}{A_i(0)}. \quad (7)$$

In the absence of a magnetic field $A_{i\uparrow}(\omega) = A_{i\downarrow}(\omega) = A_i(\omega)$ is spin-independent. A large thermopower is thus obtained for a highly asymmetric $A_i(\omega)$ with a large slope in the derivative at the Fermi energy $dA_i(\omega)/d\omega|_{\omega=0}$ and a small $A_i(0)$. As we will see below, in the Kondo regime of a double quantum dot, such an asymmetry with opposite slope for the two dots is indeed possible—in contrast to the single quantum dot case.

The physical constraints for a large thermopower can be further elucidated by relating $A_i(\omega)$ to the occupation $n_{i\sigma}$ of dot i for spin σ via the Friedel sum rule. The dot- and spin-resolved thermopower can hence be expressed as [22]

$$S_{i\sigma}(T) = -\frac{\pi\gamma T}{|e|} \cot(\pi n_{i\sigma}), \quad (8)$$

with the linear coefficient of the specific heat γ . For a single quantum dot in the Kondo regime, $n_{i\sigma} \sim 1/2$ so that $S_{i\sigma} \sim 0$ is vanishingly small [14]. Applying an external magnetic field leads to, say, $n_{i\uparrow} > 1/2$ and $n_{i\downarrow} < 1/2$. Hence, $S_{i\uparrow} > 0$ and $S_{i\downarrow} < 0$, but the total thermopower $\sum_\sigma S_{i\sigma}$ remains small [15,16]. For the double quantum dot, we have a similar situation but with the two distinct dots ($i = u$ and d) now playing the role of the spin (\uparrow and \downarrow). If there was not

an additional spin-degree of freedom for the double dot, we would even have the same situation, except for one important difference: the two contributions $i = u$ and d are now spatially separated. Hence, for a proper geometry, $S_{u\sigma} > 0$ and $S_{d\sigma} < 0$ can be even employed as the p - and n -type part of a thermoelectric device; see Fig. 1. We note that the case of an *attractive* interaction [15] is related to the magnetic field situation by a particle-hole transformation for one spin species. For the particle-hole transformed spin species, this also changes the sign of $S_{i\sigma}$; there is no cancellation in $\sum_\sigma S_{i\sigma}$.

IV. RESULTS

We now analyze the thermoelectric properties, the conductance, the thermopower or Seebeck coefficient, and the thermal conductivity, of the double quantum dot of Ref. [8]. Figure 2 summarizes the NRG results obtained for the Anderson model described by Eq. (1), with $U' = 163 \mu\text{eV}$, $U = 620 \mu\text{eV}$, $\Gamma_{Su} = 24 \mu\text{eV}$, $\Gamma_{Du} = 8 \mu\text{eV}$, $\Gamma_{Sd} = 38 \mu\text{eV}$, and $\Gamma_{Dd} = 21 \mu\text{eV}$ at $T = 25 \text{mK}$; see Eq. ([6]). Here, the one-particle energy levels ε_i are related to the gate voltages applied to the up and down quantum dot by

$$\begin{pmatrix} V_u \\ V_d \end{pmatrix} / \text{mV} = \begin{pmatrix} -1.62 & 1.74 \\ 1.26 & -3.19 \end{pmatrix} \begin{pmatrix} \varepsilon_u/U \\ \varepsilon_d/U \end{pmatrix} + \begin{pmatrix} -254.2 \\ -15.6 \end{pmatrix}. \quad (9)$$

The results for the conductance through both quantum dots (upper panel) reproduce the experimental data as illustrated in Ref. [8]. Increasing the gate voltage V_u (V_d) of the two electrodes 1,2 (3,4) in Fig. 1, the number of electrons in

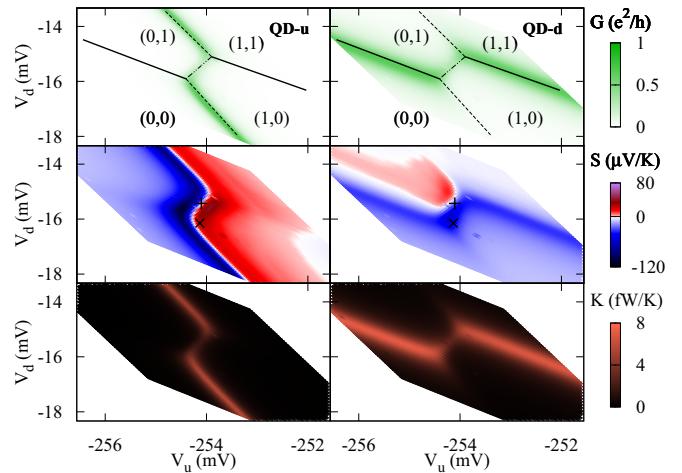


FIG. 2. (Color online) Conductance G (upper panel), Seebeck coefficient S (central panel), and thermal conductivity K^e (lower panel) for the double quantum dot of Ref. [8] as a function of the applied gate voltages V_u and V_d . The left (right) panels show the transport through the up (down) quantum dot as calculated by NRG; see text for the parameters. The Kondo resonance develops in proximity of the degeneracy line between (1,0) and (0,1) electrons in the (up, down) quantum dot, see charge stability diagram in the upper panels. Close to the maximum in the conductance, the Seebeck coefficient of the up (down) quantum dot is positive (negative), indicating charge transport of opposite sign through the two dots.

the up (down) dot is increased by one; at the degeneracy point the Coulomb blockade is lifted leading to an enhanced conductance. Due to the capacitive coupling of the dots, the gate voltages V_u and V_d do not only affect the up and down quantum dot individually, but both dots. As a consequence, instead of a square-like pattern a characteristic honeycomb structure emerges in the charge stability diagram as depicted in the upper panel of Fig. 2.

Since the Anderson impurity model describes the measured conductance very well, we are confident that the theoretical results for the thermoelectric properties correspond to the actual experimental situation. The central panels of Fig. 2 show the thermopower S_i of the two quantum dots (left and right panel, respectively). Negative (blue) values indicate the flow of negatively charged (quasi-)electrons from the hot to the cold side of an external heat gradient, and positive (red) values the flow of positively charged (quasi-)holes. At the maximal conductance (see upper panel), the thermopower is rather small. Shifting the gate voltages slightly away from this maximum, e.g., to the cross \times indicated in Fig. 2, a large thermopower develops whose values are particularly enhanced in the Kondo regime, for both quantum dots but with opposite sign.

Hence, as indicated in Fig. 1, a thermal gradient induces a current in opposite direction through the up and down quantum dot. This means that the double quantum dot constitutes a thermoelectric device. Connecting the drain electrodes of the two quantum dots, i.e., employing an even simpler design were the two drain electrodes are substituted by a bar or large quantum dot, the two source electrodes S_u and S_d in Fig. 1 act as the plus and minus source of a power supply.

To further elucidate the understanding of the large thermopower of opposite sign, we show in Fig. 3 the spectral functions of both quantum dots for the gate voltages indicated in Fig. 2. We observe that the sharp Kondo resonance of the two quantum dots is located directly below and above the Fermi level for the up and down quantum dot, respectively. Hence, the spectral functions $A_i(\omega)$ are highly asymmetric around the Fermi level. On the contrary, for a symmetric behavior

around the Fermi level (dashed line in Fig. 3), electrons and holes alike migrate from the hot to the cold side and their net current cancels, $I_i^1 \approx 0$. In general, we have such a symmetric situation on the line separating the $(0,1)$ and $(1,0)$ occupation regions (see upper panel of Fig. 2). Changing the gate voltages perpendicular to this line leads to a splitting of the u - and d -dot Kondo resonance, similar to applying a magnetic field for a single quantum dot. Note that due to its larger hybridization or tunneling rate $\Gamma_{Ld} + \Gamma_{Rd}$, the down dot has a much wider resonance and the situation is not completely symmetric. As a function of temperature, the maximum in the thermopower occurs on a temperature scale which correlates with the gate voltage and is therefore highly tunable. However, a systematic study of the temperature dependence has not been performed for the device considered here.

An efficient thermoelectric element is characterized also by a low thermal conductivity K^e , which determines the thermoelectric figure of merit defined by [14] $ZT = S^2 TG / K$. Figure 2 shows the electronic contribution to K^e determined by Eq. (4). The double dot exhibits a maximum in the thermal conductivity in correspondence of a maximum in the conductance. A large thermopower is, however, observed slightly off this maximum, where the thermal conductivity is strongly suppressed.

For the thermoelectric figure of merit $ZT = S^2 TG / (K^e + K^{ph})$ also the phononic contribution to the thermal conductivity K^{ph} has to be considered. For a reduced electronic (thermal) conductivity, this phononic contribution is dominating. In contrast to bulk materials, the thermal conductivity of nanostructures can be strongly suppressed. This is exploited in phonon engineering [23], which led to a historic breakthrough of higher figure of merit ZT achieved in nanostructured materials, e.g., for quantum dot lattices [24]. On general grounds, one can expect a suppressed phononic contribution to the thermal conductivity for nanostructures below the typical phonon mean free path. For semiconductors such as Si and GaAs, this mean free path is $0.1\text{--}1 \mu\text{m}$ [25], such that the phononic thermal conductivity is expected to be small.

In the case of the molecule instead of the quantum dot realization, the phonon contribution has been estimated in Ref. [15] to be between $K^{ph} = 0$ (best case, no phonons) and $K^{ph} = \pi^2 k_B T$ (worst case, maximal phonon contribution), resulting in $ZT \gtrsim 1$ and $ZT \sim 2 \times 10^{-2}$, respectively, for the attractive (negative) U single quantum dot. Due to the aforementioned particle-role relation, similar values for an optimal figure of merit are to be expected for our case of a double dot (or molecule) with repulsive U ; the actual ZT of the presented calculations, which are based on the experimentally realized quantum dots [6,8] and $T = 25 \text{ mK}$, instead of parameters optimizing the thermoelectric properties, are smaller. Let us also remark that comparable efficiencies can be obtained also for noninteracting quantum dots, albeit at temperatures $T \sim \mathcal{O}(\Gamma_L + \Gamma_R)$, while for the Kondo quantum dots the maximal ZT is at $T \sim \mathcal{O}(T_K) \ll \Gamma_L + \Gamma_R$.

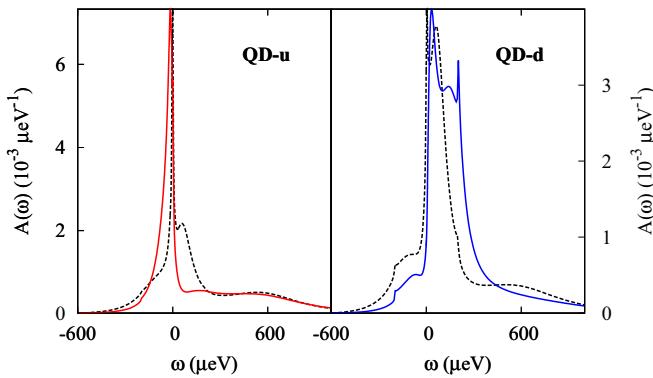


FIG. 3. (Color online) Left (right) panel: Spectral function $A_{u(d)}$ of the up (down) quantum dot for $V_u = -254.6 \text{ mV}$ and $V_d = -14.8 \text{ mV}$, corresponding to a pronounced thermopower (\times) in Fig. 2 (solid lines). The main spectral weight of the Kondo resonance is below (above) the Fermi level at $\omega = 0$. For comparison the spectral function for a reduced thermopower (+) are shown (dashed lines).

V. CONCLUSION AND OUTLOOK

We have shown that a double quantum dot in the Kondo regime can be employed as a thermoelectric power generator.

Our calculations are based on the experiment of Refs. [6] and [8] whose focus was the demonstration of Kondo correlations in the electric transport, not to optimize the thermoelectric properties. Hence, the study of different parameter regimes offers plenty of room to improve the thermopower and thermoelectric figure of merit. In fact, the relevant energy scales of the double quantum dot considered were in the μeV and mK regime, which makes these particular quantum dots unpractical for applications, except for Peltier cooling at ultralow temperatures in the mK regime, which might be of interest for basic research devices. However, reducing the size of the quantum dots from μm toward nm, the corresponding energy and temperature scales will be simply rescaled, as ZT . This can be pushed to even smaller sizes by employing molecules instead of quantum dots in molecular electronics [26]. In view of future applications, multiple quantum dots connected through a common back electrode (joint drain in Fig. 1) provide a scalable setup, in which the generated power can be harvested through wires connecting the *p*- and *n*-type quantum dots separately.

Possibly most promising is the integration of the double quantum dot thermoelectric element on computer chips. Here the complex and otherwise expensive semiconductor device fabrication with photolithographic and chemical processing is employed anyhow. At the same time, the power consumption and cooling of waste heat has become a critical issue for computer chip design. On-chip cooling, e.g., through liquid-filled microchannels is presently explored as a possible solution [27], which would involve additional technology and processing steps. Here, the proposed double quantum dot device is much simpler.

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